

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of :  
Beverley BROWN et al. : Group Art Unit.: 1621  
Serial No.: 10/580,552 : Examiner: NWAONICHA, Chukwuma O  
Filed: May 26, 2006 :  
Title: IMPROVEMENTS IN AND RELATING TO ORGANIC SEMICONDUCTING  
LAYERS

**APPEAL BRIEF**

Mail Stop: AF  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

Further to the Notice of Appeal filed on August 13, 2009, please consider the following.

The Appeal Brief fee of \$ 540.00 is filed/paid herewith.

The Commissioner is hereby authorized to charge any fees associated with this response or credit any overpayment to Deposit Account No. 13-3402.

**(i) REAL PARTY IN INTEREST**

The real party in interest is Merck Patent GMBH. See recordation at reel/frame: 017959/0229.

**(ii) RELATED APPEALS AND INTERFERENCES**

There are no known related appeals or interferences.

**(iii) STATUS OF CLAIMS**

Claims 1-28 are pending in the present application.

No claims are cancelled.

No claims are withdrawn from consideration.

No claims are allowed.

Claims 1-28 are rejected.

Claims 1-28 are on appeal.

**(iv) STATUS OF AMENDMENTS**

No amendments were filed after final.

**(v) SUMMARY OF CLAIMED SUBJECT MATTER**

Appellants' invention is directed,

in independent claim **1**, to an organic semiconducting layer formulation which contains an organic binder which has a permittivity,  $\epsilon$ , at 1,000 Hz of 3.3 or less; and a polyacene compound of Formula A (see page 2, line 19 to page 3, line 14);

in independent claim **23**, to an OFET device, which contains an organic semiconducting layer formulation, which formulation contains a compound of Formula 1; a binder (which is poly( $\alpha$ -methylstyrene), linear olefin and cycloolefin(norbornene)copolymer, poly(4-methylstyrene), polystyrene or polystyrene-co- $\alpha$ -methylstyrene); and a solvent (which is toluene, ethylcyclohexane, anisole or pxylylene) (see page 1, lines 7-13, page 5, line 10 to page 6, line 7, and page 7, lines 1-11);

in independent claim **24**, to an OFET device, which contains an organic semiconducting layer formulation, which formulation contains a compound of Formula 2; a binder (which is poly( $\alpha$ -methylstyrene), polyvinylcinnamate, or poly(4-vinylbiphenyl)); and a the solvent (which is 1,2-dichlorobenzene) (see page 1, lines 7-13, page 5, line 10 to page 6, line 7, and page 7, lines 12-21);

in independent claim **25**, to an OFET device, which contains an organic semiconducting layer formulation, which formulation contains a compound of Formula 3; a binder (which is poly( $\alpha$ -methylstyrene)); and a solvent (which is toluene) (see page 1, lines 7-13, page 5, line 10 to page 6, line 7, and page 7, line 23 to page 8, line 11);

in independent claim **26**, to a compound of Formula 3 (see page 3, lines 27-31 and page 5, line 10 to page 6, line 7); and

in independent claim **28**, to an organic semiconducting layer formulation, which contains an organic binder which has a permittivity,  $\epsilon$ , at 1,000 Hz of 3.3 or less; and a polyacene compound which is of Formula 1, Formula 2, or Formula 3 (see page 2, line 19 to page 3, line 14).

**(vi) GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL**

The ground for rejection is the rejection under 35 U.S.C. § 103, i.e., whether claims 1-28 are patentable over Brown et al. (WO 0245184 or its equivalent US 7,095,044) in view of Minakata (WO 2003016599 or its equivalent US 7,061,010).

**(vii) ARGUMENT**

The rejection under 35 U.S.C. § 103 of claim 1 and its dependent claims, i.e., whether they are patentable over Brown et al. in view of Minakata

The Office Action dated March 13, 2009 (Office Action hereinafter) rejects the claims as allegedly obvious over Brown et al. in view of Minakata.<sup>1</sup>

Brown teaches an organic semiconductor in combination with an organic binder. See column 3, lines 4-7. **Brown** teaches that: organic semi-conducting materials include “soluble” compounds (see column 4, lines 9-10), when certain polymeric semiconducting materials are “insoluble, then analogues thereof could be used” (see column 8, lines 63-65), if a preformed binder is used it may be “dissolved” together with the semiconductor (see column 9, lines 36-37), both the binder and organic semi-conductor are “dissolved” (see column 10, lines 2-3), and in the examples, a mixture of semiconductor and binder were “dissolved” in a solvent (see column 23, lines 39-40). (Emphases added.)

On the other hand, **Minakata** teaches that “crystal growth” can be controlled to form an organic semiconductor thin film “having high crystallinity,” and that the resultant organic semiconductor thin film has “superior characteristics as a semiconductor due to high crystallinity” (see column 6, lines 32-39), formed crystals tend to have polyacene molecules with their long axis perpendicular to the face of the substrate (see column 6, lines 48-51), organic semiconductor thin films having “high crystallinity” (see column 7, lines 1-4), the organic semiconductor thin films of the present invention “have superior semiconductor characteristics since these thin films are almost defect-free and have high crystallinity” (see also column 25, lines 6-10). (Emphases added.)

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<sup>1</sup> The Advisory Action dated August 11, 2009, provides no further input on why the rejections were maintained in view of the Response filed on July 13, 2009, which provides arguments and discussion of data in the application demonstrating significant unexpected advantages over the closest embodiments in both cited prior art references. The only reason (which is merely a conclusion) provided in the Advisory Action for the maintenance of the rejection is that the arguments were “not persuasive because Applicants claimed formulation and device are obvious.”

The invention of Minakata is based on the idea that pentacene in its pure, crystalline state is useful in the semiconductor layer, and said crystallinity is responsible for the superior characteristics of the product.

Consequently, according to the teaching of Minakata, the dissolving of the pentacene in the layer in any way (e.g. by mixing it with a binder) would be totally undesired and directly contrary to the teachings recited above, since it would be expected by one of ordinary skill in the art that the use of a binder would disrupt the crystalline structure of the pentacene, i.e., this is a situation of a teaching away. As such, one of ordinary skill in the art would not find it desirable or even practicable to combine the teachings of these two references. Even for this reason alone, the rejection should be reversed.

As explained in the application on page 1, line 35 to page 2, line 13, when the substituted pentacene is mixed with an organic binder, it is effectively "diluted" by the binder. Diluting the organic semiconductor by mixing it with binders disrupts the molecular order in the semiconducting layer. Hence, following the teaching of Minakata, the person skilled in the art would rather expect a reduction of the charge mobility as Minakata teaches the importance of maintaining the crystallinity of the semiconductor. Also, diluting an organic semiconductor in the channel of an OFET for example is particularly problematic, as any disruption of the orbital overlap between molecules in the immediate vicinity of the gate insulator (the first few molecular layers) is expected to reduce mobility. Electrons or holes are then forced to extend their path into the bulk of the organic semiconductor, which is undesirable. See, e.g., Minakata teaching the formation of films that are almost defect-free and crystalline, see column 25, line 8.

In view of the teachings of Brown and/or Minakata, it would be therefore totally surprising and unexpected for one of ordinary skill in the art that mixing a substituted pentacene with a binder can yield a transistor device with significantly increased charge carrier mobility.

Additionally, the present application provides unexpected results over the cited references.

Example 17 of Minakata is believed to be the closest prior art example. This example provides a crystallized 6,13-bis(triisopropylsilylethynyl)pentacene film with the triisopropylsilylethynyl group oriented perpendicular to the substrate surface and achieved a carrier mobility of  $0.12 \text{ cm}^2/\text{V}\cdot\text{s}$ . Compare this to example 12 of the present application where the same compound 6,13-bis(triisopropylsilylethynyl)pentacene was in combination

with a binder p- $\alpha$ MS and provided a mobility of  $0.433 (\pm 0.19) \text{ cm}^2/\text{V}\cdot\text{s}$ . The improvement is more than  $3 \frac{1}{2}$  in mobility value, which is significant and is completely unexpected considering Minakata teaches the importance of maintaining the crystallinity of the pentacene in the film.

Applicants also provide example 13 (comparative) which again tests the 6,13-bis(triisopropylsilylethynyl)pentacene without a binder and achieves a mobility of  $0.14 (\pm 0.14) \text{ cm}^2/\text{V}\cdot\text{s}$ , which is consistent with the findings of Minakata.

Additionally, unexpected results are provided over the examples of Brown et al. also. See, e.g., the table in Brown on columns 25 and 26. Here, 5 binders (see key to identity of binders on column 23) are tested with various semiconductor compounds (which are not pentacenes). All mobility values with these binders were in the range of  $10^{-4}$  to  $10^{-5} \text{ cm}^2/\text{V}\cdot\text{s}$  (see also claim 7 in Brown). Binders 2, 3 and 4 are also exemplified in the present application with pentacenes. See binders P-4MS, PS-co- $\alpha$ MS, and P- $\alpha$ MS respectively (see key to identity of binders on page 55). The results with these same binders in combination with a pentacene yielded results in the  $0.16$  to  $1.1 \text{ cm}^2/\text{V}\cdot\text{s}$  range (see results in the tables on pages 52 to 54 of the specification), which are several fold higher than the results obtained with these same binders and other semiconductor compounds by Brown. In view of the consistency of the results with various semiconductor compounds by Brown, it is highly unexpected to one of ordinary skill in the art that another semiconductor compound, i.e., in the present case polyacene, would yield such significantly higher mobility values.

Accordingly, this additional reason also supports the patentability of the present invention.

Additionally, applicants provide the following further comments.

In the semiconducting films of the present application (like in examples 12 and 14) the substituted pentacene compounds are not completely dissolved, but are phase-separated as small crystalline domains in the amorphous binder. In contrast, in the examples of US 7,061,010 (Minakata, Asahi Glass), as well as in the comparative examples 13 and 15 of this application, the pentacene forms a pure crystalline film as expected in view of the teachings of Minakata.

The pentacene/binder films of the present application have several additional advantages, as set forth below.

- Processing and film formation during device manufacture is much easier

because there is no need to have controlled crystallization, and the pentacene/binder composite can more easily be coated and transformed into a thin film, e.g., because the amorphous binder polymers have much better rheology than the crystalline pentacene. Also, the use of a binder allows a much broader choice of suitable solvents. This makes the pentacene/binder system, e.g., also highly suitable for printing processes.

- The uniformity of the semiconducting properties and of the device performance is better (i.e. the charge mobility values show less variation in different areas of the film). See Table 4 in the present application, where examples 12 and 14 (with the pentacene/binder film) show a much smaller relative standard deviation of the charge mobility ( $0.433 \pm 0.19$  and  $1.1 \pm 0.4$ , which is about 40-50% of the mean value), which confirms the higher film uniformity.

In contrast, the comparative examples 13 and 15 (with a pure crystalline pentacene film) show a much larger relative standard deviation ( $0.14 \pm 0.14$  and  $0.11 \pm 0.11$  which is 100% of the mean value).

- Nevertheless, the mobility in the claimed pentacene/binder film is similar or even higher than in a pure pentacene film, as shown in examples 12 and 14 and comparative examples 13 and 15.

- Another advantage is that by diluting the pentacene with the binder it is possible to save costs (compared to a pure pentacene film) because the binder material is much cheaper than the pentacene.

These advantages could not be expected from the cited prior art, since Minakata does not suggest to dilute the pentacene with a binder and Brown does not suggest to use the substituted pentacenes of formula I of the present claims.

Moreover, Minakata reports that a film of 2,3-bis(triisopropylsilylethynyl)pentacene of his example 15 (which is clearly described as being crystalline) has a mobility of only  $0.08 \text{ cm}^2/\text{V}\cdot\text{s}$ , which is lower than in most of the examples of the present application. The skilled person could not expect that by diluting the substituted pentacene of Minakata with a binder, it would be possible to even increase the mobility, and in addition to achieve further advantages like an increased uniformity. This was also not suggested by Brown, because Brown does not show a direct comparison between semiconducting films with and without binder. In particular, Brown does not suggest a possible increase of the mobility (and a much lower standard deviation of the mobility) when using a binder.

For all the foregoing, reconsideration is respectfully requested.

The rejection under 35 U.S.C. § 103 of claim 23, i.e., whether it is patentable over Brown et al. in view of Minakata

The Office Action dated March 13, 2009 (Office Action hereinafter) rejects the claims as allegedly obvious over Brown et al. in view of Minakata.<sup>2</sup>

Brown teaches an organic semiconductor in combination with an organic binder. See column 3, lines 4-7. **Brown** teaches that: organic semi-conducting materials include “soluble” compounds (see column 4, lines 9-10), when certain polymeric semiconducting materials are “insoluble, then analogues thereof could be used” (see column 8, lines 63-65), if a preformed binder is used it may be “dissolved” together with the semiconductor (see column 9, lines 36-37), both the binder and organic semi-conductor are “dissolved” (see column 10, lines 2-3), and in the examples, a mixture of semiconductor and binder were “dissolved” in a solvent (see column 23, lines 39-40). (Emphases added.)

On the other hand, **Minakata** teaches that “crystal growth” can be controlled to form an organic semiconductor thin film “having high crystallinity,” and that the resultant organic semiconductor thin film has “superior characteristics as a semiconductor due to high crystallinity” (see column 6, lines 32-39), formed crystals tend to have polyacene molecules with their long axis perpendicular to the face of the substrate (see column 6, lines 48-51), organic semiconductor thin films having “high crystallinity” (see column 7, lines 1-4), the organic semiconductor thin films of the present invention “have superior semiconductor characteristics since these thin films are almost defect-free and have high crystallinity” (see also column 25, lines 6-10). (Emphases added.)

The invention of Minakata is based on the idea that pentacene in its pure, crystalline state is useful in the semiconductor layer, and said crystallinity is responsible for the superior characteristics of the product.

Consequently, according to the teaching of Minakata, the dissolving of the pentacene

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in the layer in any way (e.g. by mixing it with a binder) would be totally undesired and directly contrary to the teachings recited above, since it would be expected by one of ordinary skill in the art that the use of a binder would disrupt the crystalline structure of the pentacene, i.e., this is a situation of a teaching away. As such, one of ordinary skill in the art would not find it desirable or even practicable to combine the teachings of these two references. Even for this reason alone, the rejection should be reversed.

As explained in the application on page 1, line 35 to page 2, line 13, when the substituted pentacene is mixed with an organic binder, it is effectively "diluted" by the binder. Diluting the organic semiconductor by mixing it with binders disrupts the molecular order in the semiconducting layer. Hence, following the teaching of Minakata, the person skilled in the art would rather expect a reduction of the charge mobility as Minakata teaches the importance of maintaining the crystallinity of the semiconductor. Also, diluting an organic semiconductor in the channel of an OFET for example is particularly problematic, as any disruption of the orbital overlap between molecules in the immediate vicinity of the gate insulator (the first few molecular layers) is expected to reduce mobility. Electrons or holes are then forced to extend their path into the bulk of the organic semiconductor, which is undesirable. See, e.g., Minakata teaching the formation of films that are almost defect-free and crystalline, see column 25, line 8.

In view of the teachings of Brown and/or Minakata, it would be therefore totally surprising and unexpected for one of ordinary skill in the art that mixing a substituted pentacene with a binder can yield a transistor device with significantly increased charge carrier mobility.

Additionally, the present application provides unexpected results over the cited references.

Example 17 of Minakata is believed to be the closest prior art example. This example provides a crystallized 6,13-bis(triisopropylsilylethynyl)pentacene film with the triisopropylsilylethynyl group oriented perpendicular to the substrate surface and achieved a carrier mobility of  $0.12 \text{ cm}^2/\text{V}\cdot\text{s}$ . Compare this to example 12 of the present application where the same compound 6,13-bis(triisopropylsilylethynyl)pentacene was in combination with a binder p- $\alpha$ MS and provided a mobility of  $0.433 (\pm 0.19) \text{ cm}^2/\text{V}\cdot\text{s}$ . The improvement is more than  $3 \frac{1}{2}$  in mobility value, which is significant and is completely unexpected considering Minakata teaches the importance of maintaining the crystallinity of the pentacene in the film.



Applicants also provide example 13 (comparative) which again tests the 6,13-bis(triisopropylsilylethynyl)pentacene without a binder and achieves a mobility of  $0.14 (\pm 0.14) \text{ cm}^2/\text{V}\cdot\text{s}$ , which is consistent with the findings of Minakata.

Additionally, unexpected results are provided over the examples of Brown et al. also. See, e.g., the table in Brown on columns 25 and 26. Here, 5 binders (see key to identity of binders on column 23) are tested with various semiconductor compounds (which are not pentacenes). All mobility values with these binders were in the range of  $10^{-4}$  to  $10^{-5} \text{ cm}^2/\text{V}\cdot\text{s}$  (see also claim 7 in Brown). Binders 2, 3 and 4 are also exemplified in the present application with pentacenes. See binders P-4MS, PS-co- $\alpha$ MS, and P- $\alpha$ MS respectively (see key to identity of binders on page 55). The results with these same binders in combination with a pentacene yielded results in the  $0.16$  to  $1.1 \text{ cm}^2/\text{V}\cdot\text{s}$  range (see results in the tables on pages 52 to 54 of the specification), which are several fold higher than the results obtained with these same binders and other semiconductor compounds by Brown. In view of the consistency of the results with various semiconductor compounds by Brown, it is highly unexpected to one of ordinary skill in the art that another semiconductor compound, i.e., in the present case polyacene, would yield such significantly higher mobility values.

Accordingly, this additional reason also supports the patentability of the present invention.

Additionally, applicants provide the following further comments.

In the semiconducting films of the present application (like in examples 12 and 14) the substituted pentacene compounds are not completely dissolved, but are phase-separated as small crystalline domains in the amorphous binder. In contrast, in the examples of US 7,061,010 (Minakata, Asahi Glass), as well as in the comparative examples 13 and 15 of this application, the pentacene forms a pure crystalline film as expected in view of the teachings of Minakata.

The pentacene/binder films of the present application have several additional advantages, as set forth below.

- Processing and film formation during device manufacture is much easier because there is no need to have controlled crystallization, and the pentacene/binder composite can more easily be coated and transformed into a thin film, e.g., because the amorphous binder polymers have much better rheology than the crystalline pentacene. Also, the use of a binder allows a much broader choice of suitable solvents. This makes the pentacene/binder

system, e.g., also highly suitable for printing processes.

- The uniformity of the semiconducting properties and of the device performance is better (i.e. the charge mobility values show less variation in different areas of the film). See Table 4 in the present application, where examples 12 and 14 (with the pentacene/binder film) show a much smaller relative standard deviation of the charge mobility ( $0.433 \pm 0.19$  and  $1.1 \pm 0.4$ , which is about 40-50% of the mean value), which confirms the higher film uniformity.

In contrast, the comparative examples 13 and 15 (with a pure crystalline pentacene film) show a much larger relative standard deviation ( $0.14 \pm 0.14$  and  $0.11 \pm 0.11$  which is 100% of the mean value).

- Nevertheless, the mobility in the claimed pentacene/binder film is similar or even higher than in a pure pentacene film, as shown in examples 12 and 14 and comparative examples 13 and 15.

- Another advantage is that by diluting the pentacene with the binder it is possible to save costs (compared to a pure pentacene film) because the binder material is much cheaper than the pentacene.

These advantages could not be expected from the cited prior art, since Minakata does not suggest to dilute the pentacene with a binder and Brown does not suggest to use the substituted pentacenes of formula I of the present claims.

Moreover, Minakata reports that a film of 2,3-bis(triisopropylsilylethynyl)pentacene of his example 15 (which is clearly described as being crystalline) has a mobility of only  $0.08 \text{ cm}^2/\text{V}\cdot\text{s}$ , which is lower than in most of the examples of the present application. The skilled person could not expect that by diluting the substituted pentacene of Minakata with a binder, it would be possible to even increase the mobility, and in addition to achieve further advantages like an increased uniformity. This was also not suggested by Brown, because Brown does not show a direct comparison between semiconducting films with and without binder. In particular, Brown does not suggest a possible increase of the mobility (and a much lower standard deviation of the mobility) when using a binder.

For all the foregoing, reconsideration is respectfully requested.

The rejection under 35 U.S.C. § 103 of claim 24, i.e., whether it is patentable over Brown et al. in view of Minakata

The Office Action dated March 13, 2009 (Office Action hereinafter) rejects the claims

as allegedly obvious over Brown et al. in view of Minakata.<sup>3</sup>

Brown teaches an organic semiconductor in combination with an organic binder. See column 3, lines 4-7. **Brown** teaches that: organic semi-conducting materials include “soluble” compounds (see column 4, lines 9-10), when certain polymeric semiconducting materials are “insoluble, then analogues thereof could be used” (see column 8, lines 63-65), if a preformed binder is used it may be “dissolved” together with the semiconductor (see column 9, lines 36-37), both the binder and organic semi-conductor are “dissolved” (see column 10, lines 2-3), and in the examples, a mixture of semiconductor and binder were “dissolved” in a solvent (see column 23, lines 39-40). (Emphases added.)

On the other hand, **Minakata** teaches that “crystal growth” can be controlled to form an organic semiconductor thin film “having high crystallinity,” and that the resultant organic semiconductor thin film has “superior characteristics as a semiconductor due to high crystallinity” (see column 6, lines 32-39), formed crystals tend to have polyacene molecules with their long axis perpendicular to the face of the substrate (see column 6, lines 48-51), organic semiconductor thin films having “high crystallinity” (see column 7, lines 1-4), the organic semiconductor thin films of the present invention “have superior semiconductor characteristics since these thin films are almost defect-free and have high crystallinity” (see also column 25, lines 6-10). (Emphases added.)

The invention of Minakata is based on the idea that pentacene in its pure, crystalline state is useful in the semiconductor layer, and said crystallinity is responsible for the superior characteristics of the product.

Consequently, according to the teaching of Minakata, the dissolving of the pentacene in the layer in any way (e.g. by mixing it with a binder) would be totally undesired and directly contrary to the teachings recited above, since it would be expected by one of ordinary skill in the art that the use of a binder would disrupt the crystalline structure of the pentacene, i.e., this is a situation of a teaching away. As such, one of ordinary skill in the art would not find it desirable or even practicable to combine the teachings of these two references. Even

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for this reason alone, the rejection should be reversed.

As explained in the application on page 1, line 35 to page 2, line 13, when the substituted pentacene is mixed with an organic binder, it is effectively "diluted" by the binder. Diluting the organic semiconductor by mixing it with binders disrupts the molecular order in the semiconducting layer. Hence, following the teaching of Minakata, the person skilled in the art would rather expect a reduction of the charge mobility as Minakata teaches the importance of maintaining the crystallinity of the semiconductor. Also, diluting an organic semiconductor in the channel of an OFET for example is particularly problematic, as any disruption of the orbital overlap between molecules in the immediate vicinity of the gate insulator (the first few molecular layers) is expected to reduce mobility. Electrons or holes are then forced to extend their path into the bulk of the organic semiconductor, which is undesirable. See, e.g., Minakata teaching the formation of films that are almost defect-free and crystalline, see column 25, line 8.

In view of the teachings of Brown and/or Minakata, it would be therefore totally surprising and unexpected for one of ordinary skill in the art that mixing a substituted pentacene with a binder can yield a transistor device with significantly increased charge carrier mobility.

Moreover, Minakata teaches polyacenes by a very broad general formula (I) depicted on column 2, lines 51-62, provides possible substituents thereon on column 2, lines 31-41 and column 2, line 63 to column 3, line 15, and provides specific embodiments thereof in the examples (see, e.g., example 9, column 15, line 35 to column 16, line 30, apparently setting forth all pentacene compounds disclosed). None of the disclosure in Minakata provides an adequate teaching or suggestion to render the compounds of Formula 2 recited in claim 24 obvious.

Additionally, the present application provides unexpected results over the cited references.

Example 17 of Minakata is believed to be the closest prior art example. This example provides a crystallized 6,13-bis(triisopropylsilylethynyl)pentacene film with the triisopropylsilylethynyl group oriented perpendicular to the substrate surface and achieved a carrier mobility of  $0.12 \text{ cm}^2/\text{V}\cdot\text{s}$ . Compare this to example 12 of the present application where the same compound 6,13-bis(triisopropylsilylethynyl)pentacene (although not a compound recited in claim 24) was in combination with a binder p- $\alpha$ MS and provided a mobility of  $0.433 (\pm 0.19) \text{ cm}^2/\text{V}\cdot\text{s}$ . The improvement is more than  $3 \frac{1}{2}$  in mobility value,

which is significant and is completely unexpected considering Minakata teaches the importance of maintaining the crystallinity of the pentacene in the film.

Applicants also provide example 13 (comparative) which again tests the 6,13-bis(triisopropylsilylethynyl)pentacene without a binder and achieves a mobility of  $0.14 (\pm 0.14) \text{ cm}^2/\text{V}\cdot\text{s}$ , which is consistent with the findings of Minakata.

Additionally, unexpected results are provided over the examples of Brown et al. also. See, e.g., the table in Brown on columns 25 and 26. Here, 5 binders (see key to identity of binders on column 23) are tested with various semiconductor compounds (which are not pentacenes). All mobility values with these binders were in the range of  $10^{-4}$  to  $10^{-5} \text{ cm}^2/\text{V}\cdot\text{s}$  (see also claim 7 in Brown). Binders 2, 3 and 4 are also exemplified in the present application with pentacenes. See binders P-4MS, PS-co- $\alpha$ MS, and P- $\alpha$ MS respectively (see key to identity of binders on page 55). The results with these same binders in combination with a pentacene yielded results in the  $0.16$  to  $1.1 \text{ cm}^2/\text{V}\cdot\text{s}$  range (see results in the tables on pages 52 to 54 of the specification, especially with the use of the compounds of example 4, which are compounds of Formula 2 recited in this claim), which are several fold higher than the results obtained with these same binders and other semiconductor compounds by Brown. In view of the consistency of the results with various semiconductor compounds by Brown, it is highly unexpected to one of ordinary skill in the art that another semiconductor compound, i.e., in the present case polyacene, would yield such significantly higher mobility values.

Accordingly, this additional reason also supports the patentability of the present invention.

Additionally, applicants provide the following further comments.

In the semiconducting films of the present application (like in examples 12 and 14) the substituted pentacene compounds are not completely dissolved, but are phase-separated as small crystalline domains in the amorphous binder. In contrast, in the examples of US 7,061,010 (Minakata, Asahi Glass), as well as in the comparative examples 13 and 15 of this application, the pentacene forms a pure crystalline film as expected in view of the teachings of Minakata.

The pentacene/binder films of the present application have several additional advantages, as set forth below.

- Processing and film formation during device manufacture is much easier because there is no need to have controlled crystallization, and the pentacene/binder composite can more easily be coated and transformed into a

thin film, e.g., because the amorphous binder polymers have much better rheology than the crystalline pentacene. Also, the use of a binder allows a much broader choice of suitable solvents. This makes the pentacene/binder system, e.g., also highly suitable for printing processes.

- The uniformity of the semiconducting properties and of the device performance is better (i.e. the charge mobility values show less variation in different areas of the film). See Table 4 in the present application, where examples 12 and 14 (with the pentacene/binder film) show a much smaller relative standard deviation of the charge mobility ( $0.433 \pm 0.19$  and  $1.1 \pm 0.4$ , which is about 40-50% of the mean value), which confirms the higher film uniformity.

In contrast, the comparative examples 13 and 15 (with a pure crystalline pentacene film) show a much larger relative standard deviation ( $0.14 \pm 0.14$  and  $0.11 \pm 0.11$  which is 100% of the mean value).

- Nevertheless, the mobility in the claimed pentacene/binder film is similar or even higher than in a pure pentacene film, as shown in examples 12 and 14 and comparative examples 13 and 15.

- Another advantage is that by diluting the pentacene with the binder it is possible to save costs (compared to a pure pentacene film) because the binder material is much cheaper than the pentacene.

These advantages could not be expected from the cited prior art, since Minakata does not suggest to dilute the pentacene with a binder and Brown does not suggest to use the substituted pentacenes of formula I of the present claims.

Moreover, Minakata reports that a film of 2,3-bis(triisopropylsilylethynyl)pentacene of his example 15 (which is clearly described as being crystalline) has a mobility of only  $0.08 \text{ cm}^2/\text{V}\cdot\text{s}$ , which is lower than in most of the examples of the present application. The skilled person could not expect that by diluting the substituted pentacene of Minakata with a binder, it would be possible to even increase the mobility, and in addition to achieve further advantages like an increased uniformity. This was also not suggested by Brown, because Brown does not show a direct comparison between semiconducting films with and without binder. In particular, Brown does not suggest a possible increase of the mobility (and a much lower standard deviation of the mobility) when using a binder.

For all the foregoing, reconsideration is respectfully requested.

The rejection under 35 U.S.C. § 103 of claim 25, i.e., whether it is patentable over Brown et al. in view of Minakata

The Office Action dated March 13, 2009 (Office Action hereinafter) rejects claim 25 as allegedly obvious over Brown et al. in view of Minakata.<sup>4</sup>

Brown teaches an organic semiconductor in combination with an organic binder. See column 3, lines 4-7. **Brown** teaches that: organic semi-conducting materials include “soluble” compounds (see column 4, lines 9-10), when certain polymeric semiconducting materials are “insoluble, then analogues thereof could be used” (see column 8, lines 63-65), if a preformed binder is used it may be “dissolved” together with the semiconductor (see column 9, lines 36-37), both the binder and organic semi-conductor are “dissolved” (see column 10, lines 2-3), and in the examples, a mixture of semiconductor and binder were “dissolved” in a solvent (see column 23, lines 39-40). (Emphases added.)

On the other hand, **Minakata** teaches that “crystal growth” can be controlled to form an organic semiconductor thin film “having high crystallinity,” and that the resultant organic semiconductor thin film has “superior characteristics as a semiconductor due to high crystallinity” (see column 6, lines 32-39), formed crystals tend to have polyacene molecules with their long axis perpendicular to the face of the substrate (see column 6, lines 48-51), organic semiconductor thin films having “high crystallinity” (see column 7, lines 1-4), the organic semiconductor thin films of the present invention “have superior semiconductor characteristics since these thin films are almost defect-free and have high crystallinity” (see also column 25, lines 6-10). (Emphases added.)

The invention of Minakata is based on the idea that pentacene in its pure, crystalline state is useful in the semiconductor layer, and said crystallinity is responsible for the superior characteristics of the product.

Consequently, according to the teaching of Minakata, the dissolving of the pentacene in the layer in any way (e.g. by mixing it with a binder) would be totally undesired and directly contrary to the teachings recited above, since it would be expected by one of ordinary

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<sup>4</sup> The Advisory Action dated August 11, 2009, provides no further input on why the rejections were maintained in view of the Response filed on July 13, 2009, which provides arguments and discussion of data in the application demonstrating significant unexpected advantages over the closest embodiments in both cited prior art references. The only reason (which is merely a conclusion) provided in the Advisory Action for the maintenance of the rejection is that the arguments were “not persuasive because Applicants claimed formulation and device are obvious.”

skill in the art that the use of a binder would disrupt the crystalline structure of the pentacene, i.e., this is a situation of a teaching away. As such, one of ordinary skill in the art would not find it desirable or even practicable to combine the teachings of these two references. Even for this reason alone, the rejection should be reversed.

As explained in the application on page 1, line 35 to page 2, line 13, when the substituted pentacene is mixed with an organic binder, it is effectively "diluted" by the binder. Diluting the organic semiconductor by mixing it with binders disrupts the molecular order in the semiconducting layer. Hence, following the teaching of Minakata, the person skilled in the art would rather expect a reduction of the charge mobility as Minakata teaches the importance of maintaining the crystallinity of the semiconductor. Also, diluting an organic semiconductor in the channel of an OFET for example is particularly problematic, as any disruption of the orbital overlap between molecules in the immediate vicinity of the gate insulator (the first few molecular layers) is expected to reduce mobility. Electrons or holes are then forced to extend their path into the bulk of the organic semiconductor, which is undesirable. See, e.g., Minakata teaching the formation of films that are almost defect-free and crystalline, see column 25, line 8.

In view of the teachings of Brown and/or Minakata, it would be therefore totally surprising and unexpected for one of ordinary skill in the art that mixing a substituted pentacene with a binder can yield a transistor device with significantly increased charge carrier mobility.

Moreover, Minakata teaches polyacenes by a very broad general formula (I) depicted on column 2, lines 51-62, provides possible substituents thereon on column 2, lines 31-41 and column 2, line 63 to column 3, line 15, and provides specific embodiments thereof in the examples (see, e.g., example 9, column 15, line 35 to column 16, line 30, apparently setting forth all pentacene compounds disclosed). None of the disclosure in Minakata provides an adequate teaching or suggestion to render the compounds of Formula 3 recited in claim 25 obvious.

Additionally, the present application provides unexpected results over the cited references.

Example 17 of Minakata is believed to be the closest prior art example. This example provides a crystallized 6,13-bis(triisopropylsilylethynyl)pentacene film with the triisopropylsilylethynyl group oriented perpendicular to the substrate surface and achieved a carrier mobility of  $0.12 \text{ cm}^2/\text{V}\cdot\text{s}$ . Compare this to example 12 of the present application



where the same compound 6,13-bis(triisopropylsilylethynyl)pentacene (although not a compound recited in claim 25) was in combination with a binder p- $\alpha$ MS and provided a mobility of  $0.433 (\pm 0.19) \text{ cm}^2/\text{V}\cdot\text{s}$ . The improvement is more than  $3 \frac{1}{2}$  in mobility value, which is significant and is completely unexpected considering Minakata teaches the importance of maintaining the crystallinity of the pentacene in the film.

Applicants also provide example 13 (comparative) which again tests the 6,13-bis(triisopropylsilylethynyl)pentacene without a binder and achieves a mobility of  $0.14 (\pm 0.14) \text{ cm}^2/\text{V}\cdot\text{s}$ , which is consistent with the findings of Minakata.

Additionally, unexpected results are provided over the examples of Brown et al. also. See, e.g., the table in Brown on columns 25 and 26. Here, 5 binders (see key to identity of binders on column 23) are tested with various semiconductor compounds (which are not pentacenes). All mobility values with these binders were in the range of  $10^{-4}$  to  $10^{-5} \text{ cm}^2/\text{V}\cdot\text{s}$  (see also claim 7 in Brown). Binders 2, 3 and 4 are also exemplified in the present application with pentacenes. See binders P-4MS, PS-co- $\alpha$ MS, and P- $\alpha$ MS respectively (see key to identity of binders on page 55). The results with these same binders in combination with a pentacene yielded results in the  $0.16$  to  $1.1 \text{ cm}^2/\text{V}\cdot\text{s}$  range (see results in the tables on pages 52 to 54 of the specification), which are several fold higher than the results obtained with these same binders and other semiconductor compounds by Brown. See also example number 26, the last row in the table on page 54, where an isomeric mixture of compounds 19 and 20 (compounds recited in claim 25, which are of Formula 3) providing a results of  $0.16 \text{ cm}^2/\text{V}\cdot\text{s}$ . In view of the consistency of the results with various semiconductor compounds by Brown, it is highly unexpected to one of ordinary skill in the art that another semiconductor compound, i.e., in the present case polyacene, would yield such significantly higher mobility values.

Accordingly, this additional reason also supports the patentability of the present invention.

Additionally, applicants provide the following further comments.

In the semiconducting films of the present application (like in examples 12 and 14) the substituted pentacene compounds are not completely dissolved, but are phase-separated as small crystalline domains in the amorphous binder. In contrast, in the examples of US 7,061,010 (Minakata, Asahi Glass), as well as in the comparative examples 13 and 15 of this application, the pentacene forms a pure crystalline film as expected in view of the teachings

of Minakata.

The pentacene/binder films of the present application have several additional advantages, as set forth below.

- Processing and film formation during device manufacture is much easier because there is no need to have controlled crystallization, and the pentacene/binder composite can more easily be coated and transformed into a thin film, e.g., because the amorphous binder polymers have much better rheology than the crystalline pentacene. Also, the use of a binder allows a much broader choice of suitable solvents. This makes the pentacene/binder system, e.g., also highly suitable for printing processes.
- The uniformity of the semiconducting properties and of the device performance is better (i.e. the charge mobility values show less variation in different areas of the film). See Table 4 in the present application, where examples 12 and 14 (with the pentacene/binder film) show a much smaller relative standard deviation of the charge mobility ( $0.433 \pm 0.19$  and  $1.1 \pm 0.4$ , which is about 40-50% of the mean value), which confirms the higher film uniformity. In contrast, the comparative examples 13 and 15 (with a pure crystalline pentacene film) show a much larger relative standard deviation ( $0.14 \pm 0.14$  and  $0.11 \pm 0.11$  which is 100% of the mean value).
- Nevertheless, the mobility in the claimed pentacene/binder film is similar or even higher than in a pure pentacene film, as shown in examples 12 and 14 and comparative examples 13 and 15.
- Another advantage is that by diluting the pentacene with the binder it is possible to save costs (compared to a pure pentacene film) because the binder material is much cheaper than the pentacene.

These advantages could not be expected from the cited prior art, since Minakata does not suggest to dilute the pentacene with a binder and Brown does not suggest to use the substituted pentacenes of formula I of the present claims.

Moreover, Minakata reports that a film of 2,3-bis(triisopropylsilyl)ethynylpentacene of his example 15 (which is clearly described as being crystalline) has a mobility of only  $0.08 \text{ cm}^2/\text{V}\cdot\text{s}$ , which is lower than in most of the examples of the present application. The skilled person could not expect that by diluting the substituted pentacene of Minakata with a binder, it would be possible to even increase the mobility, and in addition to achieve further

advantages like an increased uniformity. This was also not suggested by Brown, because Brown does not show a direct comparison between semiconducting films with and without binder. In particular, Brown does not suggest a possible increase of the mobility (and a much lower standard deviation of the mobility) when using a binder.

For all the foregoing, reconsideration is respectfully requested.

The rejection under 35 U.S.C. § 103 of claim 26, i.e., whether it is patentable over Brown et al. in view of Minakata

The Office Action dated March 13, 2009, (Office Action hereinafter), rejects claim 26 as allegedly obvious over Brown et al. in view of Minakata.<sup>5</sup>

No reason specific to this rejection is provided by the Office Action at all. As such, the Office Action failed to carry its burden in establishing unpatentability of this claim.

Accordingly, even for this reason alone, the rejection should be reversed.

Nevertheless, applicants provide the following additional comments.

Brown teaches an organic semiconductor in combination with an organic binder. This reference is completely irrelevant to claim 26, which claims a compound of Formula 3.

Minakata teaches polyacenes by a very broad general formula (I) depicted on column 2, lines 51-62, provides possible substituents thereon on column 2, lines 31-41 and column 2, line 63 to column 3, line 15, and provides specific embodiments thereof in the examples (see, e.g., example 9, column 15, line 35 to column 16, line 30, apparently setting forth all pentacene compounds disclosed). None of the disclosure in Minakata provides an adequate teaching or suggestion to render the compounds of Formula 3 of claim 26 obvious.

For all the foregoing, reconsideration is respectfully requested.

The rejection under 35 U.S.C. § 103 of claim 28 and its dependent claim, i.e., whether they are patentable over Brown et al. in view of Minakata

The Office Action dated March 13, 2009 (Office Action hereinafter) rejects the claims

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<sup>5</sup> The Advisory Action dated August 11, 2009, provides no further input on why the rejections were maintained in view of the Response filed on July 13, 2009, which provides arguments and discussion of data in the application demonstrating significant unexpected advantages over the closest embodiments in both cited prior art references. The only reason (which is merely a conclusion) provided in the Advisory Action for the maintenance of the rejection is that the arguments were "not persuasive because Applicants claimed formulation and device are obvious."

as allegedly obvious over Brown et al. in view of Minakata.<sup>6</sup>

Brown teaches an organic semiconductor in combination with an organic binder. See column 3, lines 4-7. **Brown** teaches that: organic semi-conducting materials include “soluble” compounds (see column 4, lines 9-10), when certain polymeric semiconducting materials are “insoluble, then analogues thereof could be used” (see column 8, lines 63-65), if a preformed binder is used it may be “dissolved” together with the semiconductor (see column 9, lines 36-37), both the binder and organic semi-conductor are “dissolved” (see column 10, lines 2-3), and in the examples, a mixture of semiconductor and binder were “dissolved” in a solvent (see column 23, lines 39-40). (Emphases added.)

On the other hand, **Minakata** teaches that “crystal growth” can be controlled to form an organic semiconductor thin film “having high crystallinity,” and that the resultant organic semiconductor thin film has “superior characteristics as a semiconductor due to high crystallinity” (see column 6, lines 32-39), formed crystals tend to have polyacene molecules with their long axis perpendicular to the face of the substrate (see column 6, lines 48-51), organic semiconductor thin films having “high crystallinity” (see column 7, lines 1-4), the organic semiconductor thin films of the present invention “have superior semiconductor characteristics since these thin films are almost defect-free and have high crystallinity” (see also column 25, lines 6-10). (Emphases added.)

The invention of Minakata is based on the idea that pentacene in its pure, crystalline state is useful in the semiconductor layer, and said crystallinity is responsible for the superior characteristics of the product.

Consequently, according to the teaching of Minakata, the dissolving of the pentacene in the layer in any way (e.g. by mixing it with a binder) would be totally undesired and directly contrary to the teachings recited above, since it would be expected by one of ordinary skill in the art that the use of a binder would disrupt the crystalline structure of the pentacene, i.e., this is a situation of a teaching away. As such, one of ordinary skill in the art would not find it desirable or even practicable to combine the teachings of these two references. Even

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<sup>6</sup> The Advisory Action dated August 11, 2009, provides no further input on why the rejections were maintained in view of the Response filed on July 13, 2009, which provides arguments and discussion of data in the application demonstrating significant unexpected advantages over the closest embodiments in both cited prior art references. The only reason (which is merely a conclusion) provided in the Advisory Action for the maintenance of the rejection is that the arguments were “not persuasive because Applicants claimed formulation and device are obvious.”

for this reason alone, the rejection should be reversed.

As explained in the application on page 1, line 35 to page 2, line 13, when the substituted pentacene is mixed with an organic binder, it is effectively "diluted" by the binder. Diluting the organic semiconductor by mixing it with binders disrupts the molecular order in the semiconducting layer. Hence, following the teaching of Minakata, the person skilled in the art would rather expect a reduction of the charge mobility as Minakata teaches the importance of maintaining the crystallinity of the semiconductor. Also, diluting an organic semiconductor in the channel of an OFET for example is particularly problematic, as any disruption of the orbital overlap between molecules in the immediate vicinity of the gate insulator (the first few molecular layers) is expected to reduce mobility. Electrons or holes are then forced to extend their path into the bulk of the organic semiconductor, which is undesirable. See, e.g., Minakata teaching the formation of films that are almost defect-free and crystalline, see column 25, line 8.

In view of the teachings of Brown and/or Minakata, it would be therefore totally surprising and unexpected for one of ordinary skill in the art that mixing a substituted pentacene with a binder can yield a transistor device with significantly increased charge carrier mobility.

Additionally, the present application provides unexpected results over the cited references.

Example 17 of Minakata is believed to be the closest prior art example. This example provides a crystallized 6,13-bis(triisopropylsilylethynyl)pentacene film with the triisopropylsilylethynyl group oriented perpendicular to the substrate surface and achieved a carrier mobility of  $0.12 \text{ cm}^2/\text{V}\cdot\text{s}$ . Compare this to example 12 of the present application where the same compound 6,13-bis(triisopropylsilylethynyl)pentacene was in combination with a binder p- $\alpha$ MS and provided a mobility of  $0.433 (\pm 0.19) \text{ cm}^2/\text{V}\cdot\text{s}$ . The improvement is more than  $3 \frac{1}{2}$  in mobility value, which is significant and is completely unexpected considering Minakata teaches the importance of maintaining the crystallinity of the pentacene in the film.

Applicants also provide example 13 (comparative) which again tests the 6,13-bis(triisopropylsilylethynyl)pentacene without a binder and achieves a mobility of  $0.14 (\pm 0.14) \text{ cm}^2/\text{V}\cdot\text{s}$ , which is consistent with the findings of Minakata.

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binders on column 23) are tested with various semiconductor compounds (which are not pentacenes). All mobility values with these binders were in the range of  $10^{-4}$  to  $10^{-5}$   $\text{cm}^2/\text{V}\cdot\text{s}$  (see also claim 7 in Brown). Binders 2, 3 and 4 are also exemplified in the present application with pentacenes. See binders P-4MS, PS-co- $\alpha$ MS, and P- $\alpha$ MS respectively (see key to identity of binders on page 55). The results with these same binders in combination with a pentacene yielded results in the 0.16 to 1.1  $\text{cm}^2/\text{V}\cdot\text{s}$  range (see results in the tables on pages 52 to 54 of the specification), which are several fold higher than the results obtained with these same binders and other semiconductor compounds by Brown. In view of the consistency of the results with various semiconductor compounds by Brown, it is highly unexpected to one of ordinary skill in the art that another semiconductor compound, i.e., in the present case polyacene, would yield such significantly higher mobility values.

Accordingly, this additional reason also supports the patentability of the present invention.

Additionally, applicants provide the following further comments.

In the semiconducting films of the present application (like in examples 12 and 14) the substituted pentacene compounds are not completely dissolved, but are phase-separated as small crystalline domains in the amorphous binder. In contrast, in the examples of US 7,061,010 (Minakata, Asahi Glass), as well as in the comparative examples 13 and 15 of this application, the pentacene forms a pure crystalline film as expected in view of the teachings of Minakata.

The pentacene/binder films of the present application have several additional advantages, as set forth below.

- Processing and film formation during device manufacture is much easier because there is no need to have controlled crystallization, and the pentacene/binder composite can more easily be coated and transformed into a thin film, e.g., because the amorphous binder polymers have much better rheology than the crystalline pentacene. Also, the use of a binder allows a much broader choice of suitable solvents. This makes the pentacene/binder system, e.g., also highly suitable for printing processes.
- The uniformity of the semiconducting properties and of the device performance is better (i.e. the charge mobility values show less variation in different areas of the film). See Table 4 in the present application, where examples 12 and 14 (with the pentacene/binder film) show a much smaller relative

standard deviation of the charge mobility ( $0.433 \pm 0.19$  and  $1.1 \pm 0.4$ , which is about 40-50% of the mean value), which confirms the higher film uniformity.

In contrast, the comparative examples 13 and 15 (with a pure crystalline pentacene film) show a much larger relative standard deviation ( $0.14 \pm 0.14$  and  $0.11 \pm 0.11$  which is 100% of the mean value).

- Nevertheless, the mobility in the claimed pentacene/binder film is similar or even higher than in a pure pentacene film, as shown in examples 12 and 14 and comparative examples 13 and 15.

- Another advantage is that by diluting the pentacene with the binder it is possible to save costs (compared to a pure pentacene film) because the binder material is much cheaper than the pentacene.

These advantages could not be expected from the cited prior art, since Minakata does not suggest to dilute the pentacene with a binder and Brown does not suggest to use the substituted pentacenes of formula I of the present claims.

Moreover, Minakata reports that a film of 2,3-bis(triisopropylsilyl)ethynylpentacene of his example 15 (which is clearly described as being crystalline) has a mobility of only  $0.08 \text{ cm}^2/\text{V}\cdot\text{s}$ , which is lower than in most of the examples of the present application. The skilled person could not expect that by diluting the substituted pentacene of Minakata with a binder, it would be possible to even increase the mobility, and in addition to achieve further advantages like an increased uniformity. This was also not suggested by Brown, because Brown does not show a direct comparison between semiconducting films with and without binder. In particular, Brown does not suggest a possible increase of the mobility (and a much lower standard deviation of the mobility) when using a binder.

For all the foregoing, reconsideration is respectfully requested.

Reversal of all the rejections is respectfully and courteously requested.

Respectfully submitted,

/Csaba Henter/

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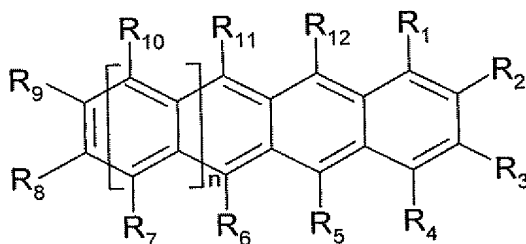
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### (viii) CLAIMS APPENDIX

1. An organic semiconducting layer formulation, comprising an organic binder which has a permittivity,  $\epsilon$ , at 1,000 Hz of 3.3 or less; and a polyacene compound of Formula A:



Formula A

wherein:

each of  $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_4$ ,  $R_5$ ,  $R_6$ ,  $R_7$ ,  $R_8$ ,  $R_9$ ,  $R_{10}$ ,  $R_{11}$  and  $R_{12}$ , which may be the same or different, independently represents hydrogen; an optionally substituted  $C_1$ - $C_{40}$  carbyl or hydrocarbyl group; an optionally substituted  $C_1$ - $C_{40}$  alkoxy group; an optionally substituted  $C_6$ - $C_{40}$  aryloxy group; an optionally substituted  $C_7$ - $C_{40}$  alkylaryloxy group; an optionally substituted  $C_2$ - $C_{40}$  alkoxycarbonyl group; an optionally substituted  $C_7$ - $C_{40}$  aryloxycarbonyl group; a cyano group ( $-CN$ ); a carbamoyl group ( $-C(=O)NH_2$ ); a haloformyl group ( $-C(=O)-X$ , wherein  $X$  represents a halogen atom); a formyl group ( $-C(=O)-H$ ); an isocyano group; an isocyanate group; a thiocyanate group or a thioisocyanate group; an optionally substituted amino group; a hydroxy group; a nitro group; a  $CF_3$  group; a halogen group; or an optionally substituted silyl group;

wherein independently each pair of  $R_2$  and  $R_3$  and/or  $R_8$  and  $R_9$ , may be cross-bridged to form a  $C_4$ - $C_{40}$  saturated or unsaturated ring, which saturated or unsaturated ring may be intervened by an oxygen atom, a sulphur atom or a group shown by formula  $-N(R_a)-$  (wherein  $R_a$  is a hydrogen atom or an optionally substituted hydrocarbon group), or may optionally be substituted;

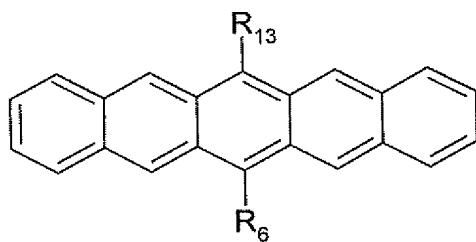
wherein one or more of the carbon atoms of the polyacene skeleton may optionally be

substituted by a N, P, As, O, S, Se or Te atom; and wherein independently any two or more of the substituents  $R_1$ - $R_{12}$  which are located on adjacent ring positions of the polyacene may, together, optionally constitute a further  $C_4$ - $C_{40}$  saturated or unsaturated ring optionally interrupted by O, S or  $-N(R_a)$  where  $R_a$  is as defined above) or an aromatic ring system, fused to the polyacene;

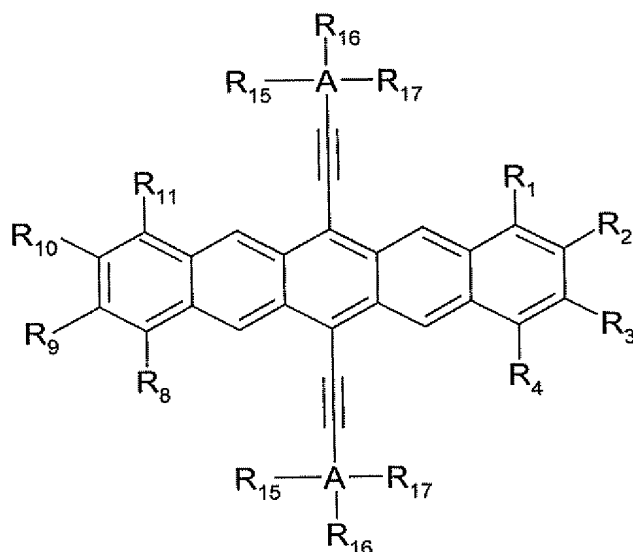
at least one of  $R_1$  to  $R_{12}$  is an optionally substituted  $C_2$ - $C_{40}$  hydrocarbyl group that is a saturated or unsaturated acyclic group, or a saturated or unsaturated cyclic group, and

$n$  is 0, 1, 2, 3 or 4.

2. An organic semiconducting layer formulation as claimed in claim 1, wherein the polyacene compound is a compound of formula B or 8 or an isomer thereof



Formula B



Formula 8

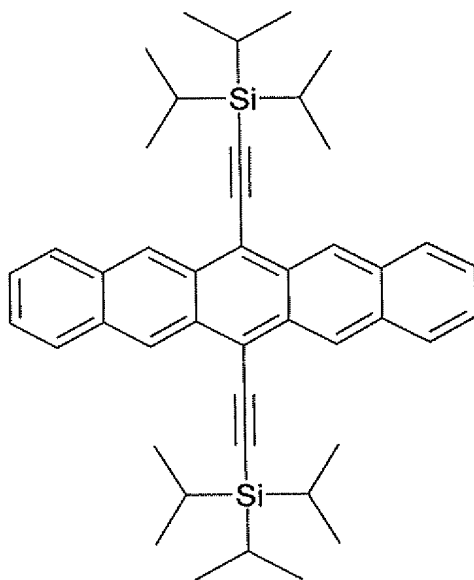
wherein,  $R_6$  and  $R_{13}$  in the compound of formula B and  $R_1, R_2, R_3, R_4, R_8, R_9, R_{10}, R_{11}, R_{15}, R_{16}$ , and  $R_{17}$  in the compound of formula 8 are each independently the same or different and each independently represents: H; an optionally substituted  $C_1$ - $C_{40}$  carbyl or hydrocarbyl group; an optionally substituted  $C_1$ - $C_{40}$  alkoxy group; an optionally substituted  $C_6$ - $C_{40}$  aryloxy group; an optionally substituted  $C_7$ - $C_{40}$  alkylaryloxy group; an optionally substituted  $C_2$ - $C_{40}$  alkoxycarbonyl group; an optionally substituted  $C_7$ - $C_{40}$  aryloxycarbonyl group; a cyano group ( $-CN$ ); a carbamoyl group ( $-C(=O)NH_2$ ); a haloformyl group ( $-C(=O)-X$ , wherein  $X$  represents a halogen atom); a formyl group ( $-C(=O)-H$ ); an isocyano group; an isocyanate group; a thiocyanate group or a thioisocyanate group; an optionally substituted amino group; a hydroxy group; a nitro group; a  $CF_3$  group; a halogen group; or an optionally substituted silyl group; and wherein independently each pair of  $R_1$  and  $R_2$ ,  $R_2$  and  $R_3$ ,  $R_3$  and  $R_4$ ,  $R_8$  and  $R_9$ ,  $R_9$  and  $R_{10}$ ,  $R_{10}$  and  $R_{11}$ ,  $R_{15}$  and  $R_{16}$  and  $R_{16}$  and  $R_{17}$  may be cross-bridged with each other to form a  $C_4$ - $C_{40}$  saturated or unsaturated ring, which saturated or unsaturated ring may be intervened by an oxygen atom, a sulphur atom or a group shown by formula:  $-N(R_a)-$  (wherein  $R_a$  is a hydrogen atom or a hydrocarbon group), or may optionally be substituted; and wherein  $A$  represents Silicon or Germanium.

3. An organic semiconducting layer formulation as claimed in claim 1, wherein  $n$  is 0 or 2.

4. An organic semiconducting layer formulation as claimed in claim 3, wherein n is 2.

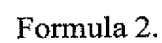
5. An organic semiconducting layer formulation as claimed in claim 1, wherein two or more of  $R_1$  to  $R_{12}$  are optionally substituted  $C_1$ - $C_{40}$  hydrocarbyl groups, each of which is a saturated or unsaturated acyclic group, or a saturated or unsaturated cyclic group.

6. An organic semiconducting layer formulation as claimed in claim 28, wherein the polyacene compound is 6, 13-bis(triisopropylsilyl)ethynyl)pentacene of Formula 1,



Formula 1.

7. An organic semiconducting layer formulation as claimed in claim 28, wherein the polyacene compound is 2,3,9,10-tetramethyl,6,13-bis (triisopropylsilyl)ethynyl)pentacene of Formula 2:



Chemical structure of a fluorinated pentacene derivative. The central pentacene core is substituted at the 1 and 8 positions with tert-butyl groups via ethynyl linkages. The 2 and 7 positions are substituted with fluorine atoms, labeled  $(F)_m$  and  $(F)_n$  respectively.

### Formula 3

wherein n and m are each independently 0, 1, 2, 3 or 4.

9. An organic semiconducting layer formulation as claimed in claim 1, wherein the organic binder resin has a permittivity at 1,000 Hz of less than 3.0.

10. An organic semiconducting layer formulation as claimed in claim 10, wherein the organic binder resin has a permittivity at 1,000 Hz greater than 1.7.

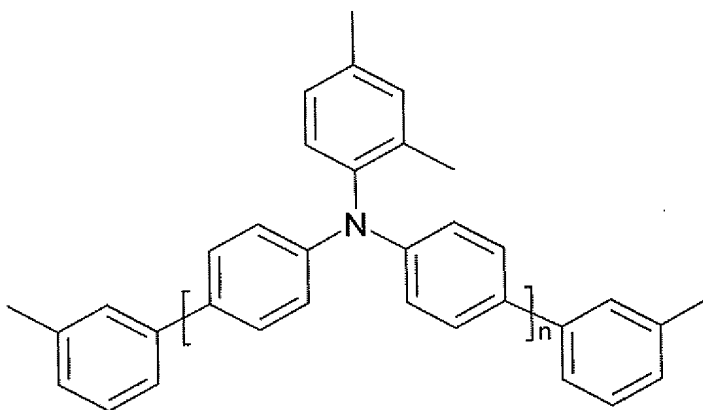
11. An organic semiconducting layer formulation as claimed in claim 1, wherein the organic binder resin is an insulating binder.

12. An organic semiconducting layer formulation as claimed in claim 11, wherein the insulating binder is poly( $\alpha$ -methylstyrene), polyvinylcinnamate, poly(4-vinylbiphenyl), poly(4-methylstyrene) or linear olefin and cycloolefin(norbornene)copolymer.

13. An organic semiconducting layer formulation as claimed in claim 1, wherein the organic binder resin is a semiconductor binder.

14. An organic semiconducting layer formulation as claimed in claim 13, wherein the semiconductor binder comprises a number average molecular weight ( $M_n$ ) of at least 1500-2000.

15. An organic semiconducting layer formulation as claimed in claim 13, wherein the semiconductor binder is poly(9-vinylcarbazole) or a triarylamine compound of the following formula



wherein  $n=10.7$ .

16. An organic semiconducting layer formulation as claimed in claim 1, wherein

the formulation further comprises a solvent.

17. An organic semiconducting layer formulation as claimed in claim 16, wherein the solvent is xylene(s), toluene, tetralin or odichlorobenzene.

18. An organic semiconducting layer formulation as claimed in claim 1, wherein the ratio of polyacence compound to binder is 20:1 to 1:20 by weight.

19. An organic semiconducting layer formulation as claimed in claim 1, which has a solids content of 0.1 to 10% by weight.

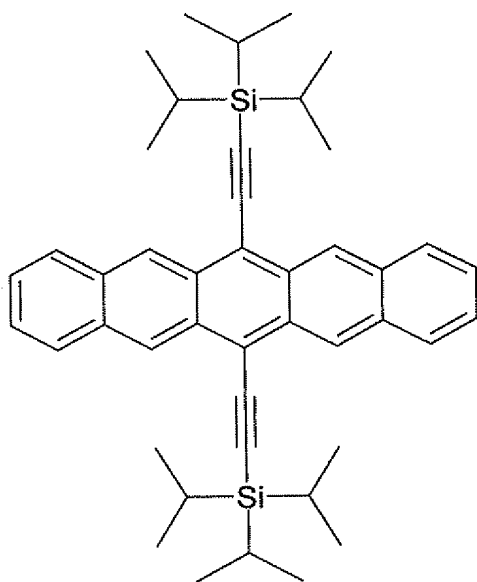
20. A process for preparing an organic semiconducting layer formulation as claimed in claim 1, comprising (i) depositing on a substrate a liquid layer of a mixture which comprises the polyacene compound, the organic binder resin or precursor thereof and optionally a solvent, and (ii) forming from the liquid layer a solid layer which is the organic semiconducting layer.

21. In an electronic device, wherein the improvement comprises the presence of an organic semiconducting layer formulation as claimed in claim 1 in said electronic device.

22. A field effect transistor (FET), organic light emitting diode (OLED), photodetector, chemical detector, photovoltaic cell (PVs), capacitor sensor, logic circuit, display or memory device, comprising an organic semiconducting layer formulation as claimed in claim 1.

23. An OFET device, comprising an organic semiconducting layer formulation, wherein the organic semiconducting layer formulation comprises:

- a compound of Formula 1;
- a binder; and
- solvent,

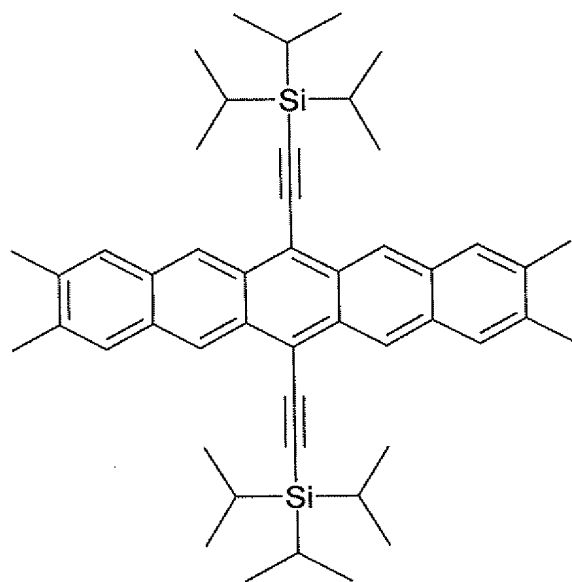


Formula 1

wherein the binder is poly( $\alpha$ -methylstyrene), linear olefin and cycloolefin(norbornene)copolymer, poly(4-methylstyrene), polystyrene or polystyrene-co- $\alpha$ -methylstyrene; and the solvent is toluene, ethylcyclohexane, anisole or pxylene.

24. An OFET device, comprising an organic semiconducting layer formulation, wherein the organic semiconducting layer formulation comprises:
- a compound of Formula 2;
  - a binder; and
  - solvent,

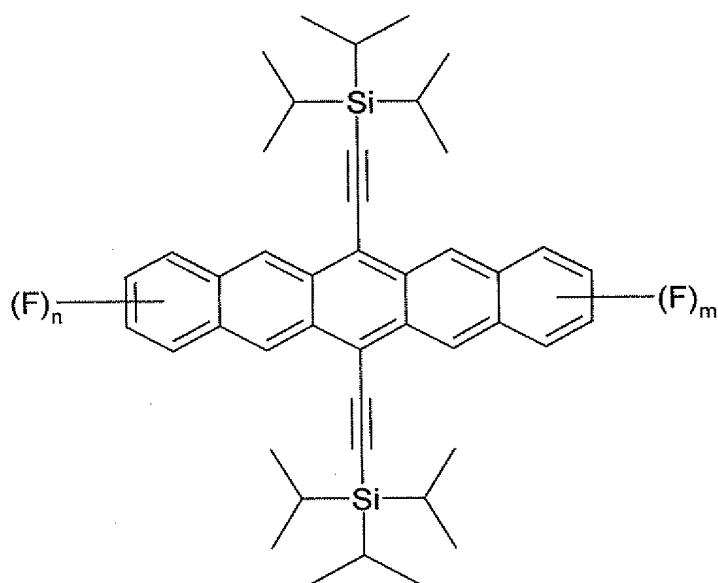




Formula 2

wherein the binder is poly( $\alpha$ -methylstyrene), polyvinylcinnamate, or poly(4-vinylbiphenyl); and the solvent is 1,2-dichlorobenzene.

25. An OFET device, comprising an organic semiconducting layer formulation, wherein the organic semiconducting layer comprises:
- a compound of Formula 3;
  - a binder; and
  - a solvent,

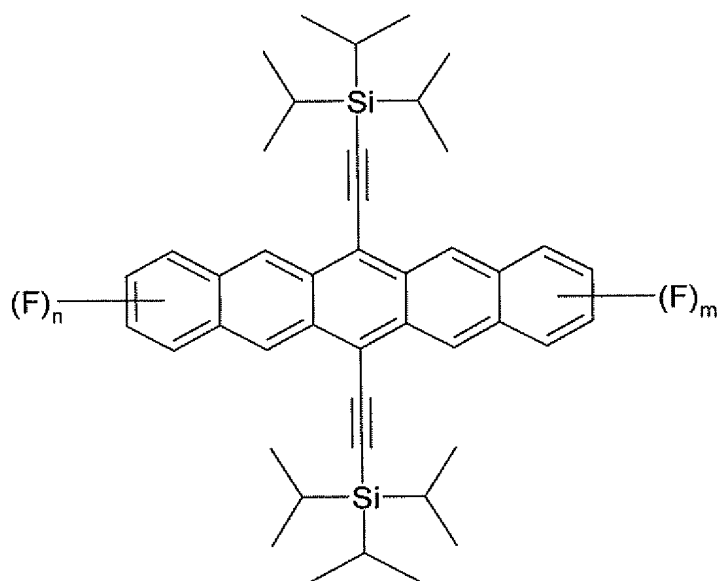


Formula (3)

wherein :

n and m are each independently 0, 1, 2, 3 or 4; the binder is poly( $\alpha$ -methylstyrene); and the solvent is toluene.

26. A compound of Formula 3



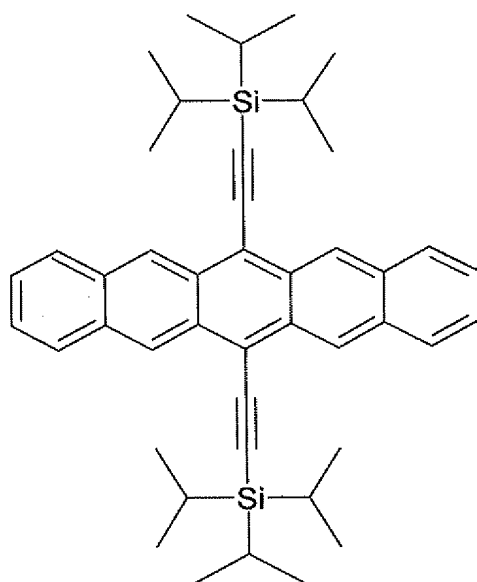
Formula (3)

wherein n and m are each independently 1 or 3.

27. An organic semiconducting layer formulation as claimed in claim 1, wherein the halogen group is Cl, Br or F.

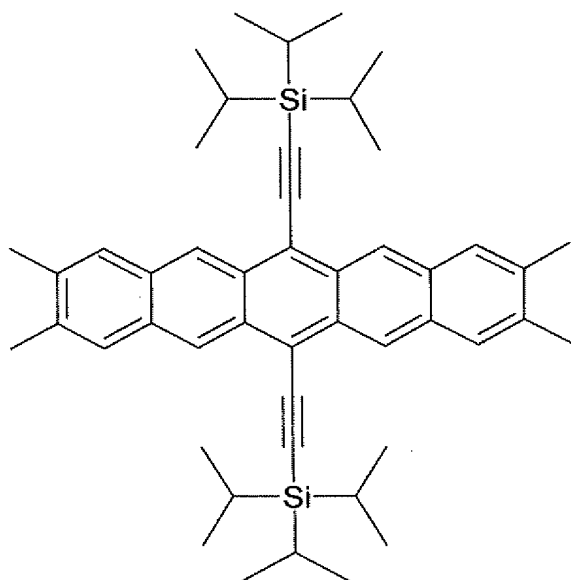
28. An organic semiconducting layer formulation, comprising an organic binder which has a permittivity,  $\epsilon$ , at 1,000 Hz of 3.3 or less; and a polyacene compound which is

a) 6, 13-bis(triisopropylsilyl)ethynyl)pentacene of Formula 1,



Formula 1;

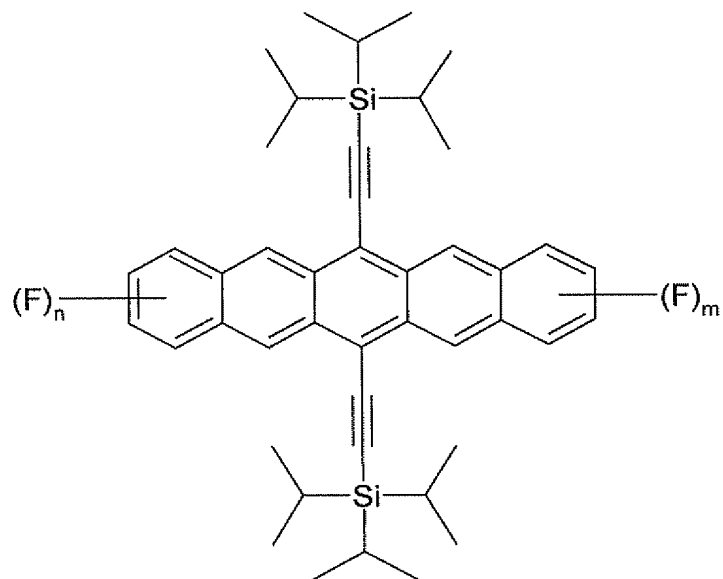
b) 2,3,9,10-tetramethyl-6,13-bis (triisopropylsilyl)ethynyl)pentacene of Formula 2:



Formula 2;

or

c) of Formula 3:



Formula 3

wherein n and m are each independently 0, 1, 2, 3 or 4.

**(ix) EVIDENCE APPENDIX**

None

**(x) RELATED PROCEEDINGS APPENDIX**

None